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PROJECT FOG DROPS

INVESTIGATION OF WARM FOG PROPERTIES AND FOG MODIFICATION CONCEPTS

by James E. Jiusto

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ABSTRACT

Considerable fog data were examined in order to quantitatively model the structural, dynamic, and microphysical characteristics of warm fog. Laboratory experiments were conducted to determine the effect of surface active materials (fatty alcohol monolayers and ionic surfactants) on the growth and evaporation rate of droplets. These models and experimental findings were applied to the evaluation of various warm fog modification concepts. Regions of high frequency of supercooled fog in the United States were delineated and pertinent fog control considerations mentioned.

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1. INTRODUCTION

Despite improved aircraft landing aids at airports, the occurrence of fog continues to hamper flight schedules. Cloud ceilings and ground visibilities less than approximately 300 feet and 0.75 mile (ILS minimums) bring air transportation to a standstill. Fog, which is the most likely weather phenomenon to test these conditions, may persist for a few hours to several days. The mean number of days with fog along the west coast is 60 per year.

Efforts to achieve a fog dispersal capability, which would have obvious safety and economic advantages, have occupied the attention of atmospheric physicists for at least two decades. The ability to dissipate thin supercooled clouds and fog has now become reality. Modification of the more prevalent "warm" fog (i.e. warmer than $0^{\circ}C$) on a practical basis has yet to be achieved. Part of the difficulty stems from insufficient knowledge of the basic structure of fog and of the chemical, physical, and electrical properties of fog droplets.

Recognizing this need, the Office of Aeronautical Research (NASA) authorized this Laboratory to undertake an investigation of warm fog properties and possible fog modification concepts. The first year's program has emphasized analytic and laboratory work on:

1. Models of the micro- and macroscopic properties of warm fogs.

2. The characteristics of aerosol droplets and means of favorably altering these properties.

3. The construction of apparatus for simulating certain fog conditions.

4. Experimental alteration of the growth or evaporation rate of otherwise stable aerosol droplets.

5. Formulation of fog modification concepts based on the above findings, as well as a review of other possible techniques.

6. Assessment of the supercooled fog problem in the United States and specification of the geographic areas where an operational seeding program might be practical.

II. SUMMARY OF PROGRAM ACCOMPLISHMENTS

1.   The fog types that affect airports in the United States were identified
and grouped into three categories: radiation fog and advection fog (the so-called
air mass fogs), and frontal fog. Structural fog models (Table II) were formulated
for radiation fog and advection fog to provide working numbers for assessing
the fog problem and for the evaluation of fog modification concepts.

2.   Visibility within fog depends largely on liquid-water-content distribution,
drop size distribution and droplet concentration. Analysis of sparse liquid
water content data suggested two fog features of potential importance in fog
control:

    a. Approximately 80% of the fog water in representative radiation
fogs consists of drops with diameters from 7 to 25 microns, and for typical
advection fogs, diameters from 8 to 35 microns. Reasonable approximations
of fog drop-size distributions can be obtained with Best's (1951) empirical
expression

$$1 - F = exp\left(-x/a\right)^{b}$$

where F is the fraction of liquid water comprised of drops of diameter less
than x; b is approximately 3.3; and a is related to liquid water content ω by

$$\omega = 1.1 \times 10^{-3} a^{1.79}$$

    b. The quantity of water in the lowest 100 m of fogs, the most
important height interval from the standpoint of landing aircraft, is relatively
modest (approximately 20 to 40 grams per 100 m x 1 m^{2} column) and conservative.
For example the water in the lowest 100 m of two markedly different fogs
varied by only a factor of two -- this despite the fact that the extremely dense
sea fog contained about 19 times as much total condensed water as the typical
inland fog investigated.

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3.　　A mathematical model of fog was needed to specify the distributions in space of temperature, water vapor, and liquid water, and also to define mechanisms and energy exchanges operative in the life cycle of warm fogs. Progress was made with a model, based on the work of Rodhe (1962), that expresses liquid water as a function of the temperature distribution in fog.

Rodhe's basic expression accounts for fog formation when mixing of moist airmasses of different temperature is the causative mechanism. This formulation will accommodate the effect of radiational cooling, which in some fogs appears necessary to explain the liquid water concentrations involved. Our work suggests that the nature and concentration of hygroscopic nuclei (plentiful in metropolitan or ocean coastal areas) also must be included, as well as two-dimensional considerations, to obtain a representative fog model.

4.　　While much experimental work has been done in the past on droplet evaporation, relatively few experiments have been conducted on droplet growth in a slightly supersaturated environment typical of fogs and clouds. A chemical-gradient cloud-diffusion chamber was constructed for this latter purpose, and techniques developed for measuring the growth rate of individual droplets.

5.　　With the above instrument, we were able to verify the theoretical prediction that a surface-active monolayer (hexadecanol, octadecanol) will retard the growth rate, dm/dt, of water drops by a factor as large as 10^2 to 10^4. It was found that the form of the governing growth-rate equation, as given by Eisner, Quince and Slack (1960) for evaporating drops, is generally valid:

$$\frac{dm}{dt} = \frac{4\pi MD}{RT}\left(fP_T - P_\theta\right)\frac{r}{1 + D/r\,r\alpha}$$

(See section III - 5B for an explanation of equation symbols.)

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Preliminary experiments indicated that condensation nuclei (NaCl) could be partially deactivated with monolayers.

6.   Monolayer stabilized drops have important implications for fog control. If effective means can be found for treating a portion of the drop population so that these drops would not compete for the available moisture, the remaining drops might then grow sufficiently large to precipitate and thereby improve visibility within the fog.   The same reasoning applies to selective deactivation of condensation nuclei prior to fog formation.   Another possible application relates to frontal fogs.   This type of fog is caused by rain falling into colder, sub-saturated air, whereupon partial evaporation of the rain increases the ambient dewpoint until fog is formed.   Conceivably, the rain drops might be stabilized with monolayers so as to inhibit evaporation and fog formation.

In the potential applications above, the sizeable task of effectively treating the fog, nuclei, or rain drops is as yet an undetermined factor.

7.   Ionic surfactants have been considered by numerous investigators in terms of their possible use for increasing diffusional growth of droplets and, more commonly, for increasing collision rates (collection efficiencies) of cloud droplets.   Laboratory experiments conducted on this project indicate that these surfactants, which tend to concentrate ions of one sign at a droplet's surface, have no significant effect on diffusional growth or evaporation rate of droplets.   To date, experiments to examine collision rates of droplets treated with surfactants have been inconclusive.

8.   A growing awareness of the importance of condensation nuclei concentration on cloud stability, fog formation, and visibility characteristics suggests the need for routine measurements of this variable at the slight supersaturations (less than a few tenths of one percent) characteristic of fogs.

9.   A review of fog modification efforts indicates that alteration of droplet growth by chemical and by electrical means warrants continued investigation. The only proven method for reasonable effective dissipation of warm fog -- Fido-type air heating -- could benefit from recent engineering advances in burner and fuel technology; past objections (expense, complexity, safety hazard) to the technique would probably still persist.

10.   A survey was made of supercooled fog frequency in the United States which showed that the Pacific Northwest is the most affected region. Certain valley airports in that area have recorded from 46 to 213 hours of supercooled fog in an average winter season. The few degree proximity to 0° C of most of these fogs would dictate the use of dry ice in seeding programs and also seeding methods that maximize dry ice - air interaction time.

III. TECHNICAL DISCUSSION

1. Fog Classification

Fog can form in one of two ways -- by ambient air cooling to its dew-point (air mass fogs) or by the moisture content of the air increasing until saturated conditions are attained (frontal fogs). Mixing of two air parcels is sometimes considered a third way to form fog, although mixing is actually a combination of heat and moisture exchange. There are numerous moisture transport and cooling mechanisms in the free atmosphere by which fog can form, each mechanism being associated with a generic fog type.

The fog classification system employed in our study is based on the classification introduced by Willett (1928) and subsequently modified by Byers (1959). A brief description of the various fog types and the classification system is given in Appendix A. Owing to the limited amount of information on fog structure and to simplify the problem in terms of our project interests, available air mass fog data were lumped into two broad categories -- advection fog and radiation fog. As indicated in Table I, advection fogs, which commonly occur in coastal regions, encompass those fog types in which cooling is caused primarily by the transport of warm air over a cold surface; radiation fogs, which commonly occur inland, comprise those fogs in which radiation constitutes the principal cooling mechanism. More frequently than not, both cooling mechanisms are operative, with one process being dominant.

Table I Fog Classification[*]

A. Advection (Coastal) Fog	B. Radiation (Inland) Fog
1. land and sea breeze fog	1. ground fog
2. sea fog	2. high inversion fog
3. tropical air (over water) fog	3. advection-radiation fog
	4. upslope fog
	5. tropical air (over land) fog

Air mass fogs, which occur more frequently than frontal fogs, are of major importance on this program. Frontal fogs are associated with active precipitation and an increase in ambient dew point in the vicinity of fronts.

Ice fogs and "arctic sea smoke" have been deleted from the study since the former occur only at polar latitudes and the latter only over open water.

2. Fog Structure -- Physical Models

A reasonably comprehensive literature survey was conducted in order to construct physical or structural models of radiation and advection fogs as herein defined. Despite the considerable research interest in fog phenomena for some three decades, the amount of consistent data on fog structure is generally too sparse for statistical treatment. The deficiency stems partly from inadequate instrumentation, especially with respect to measurements aloft, partly from the lack of standardization of measured quantities (for example, the "average" size of fog drops may, depending on the investigator, represent the arithmetic or linear mean, volume mean, area mean, median, mode etc), and partly because of the variability in fog characteristics depending on season, locale, and fog type.

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[*]See Appendix A for explanation of fog types. Note that in our classification the coastal fogs initially form over water and subsequently may be transported inland whereas the inland fogs form over a continental area.

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While statistically valid measures of pertinent fog parameters are not available, approximately 100 references (of which the most useful were listed in Progress Report 1) enabled the specification of representative fog values. These values, which are presented in Table II, along with Figures 1 and 2, constitute our tentative working models of radiation and advection fogs.

Table II Physical Fog Models

Fog Parameters at the Surface	Radiation (inland) fog	Advection (coastal) fog
1. Average Drop Dia.	10μ	20μ
2. Typical drop size range	$5-35\mu$	$7-65\mu$
3. Liquid water content	110 mg/m^3	170 mg/m^3
4. Droplet concentration	200 cm^{-3}	40 cm^{-3}
5. Vertical depth of fog		
a. typical	100 m	200 m
b. severe	300 m	600 m
6. Horizontal visibility	100 m	300 m
7. Nuclei		
a. size	$0.08 - 0.8\mu$	0.5μ and greater
b. type	combustion products	chlorides and nitrates

From the standpoint of fog modification, the liquid water content w and its associated vertical distribution is one of the more important characteristics of fog. Figure 1 shows three measured w profiles obtained in (1) an inland fog (Nikandrov 1960), (2) a dense coastal advection-radiation fog (Okita 1962), and (3) a dense sea fog (Hanajima 1945). These three profiles were used (see next page) in assessments of the fog problem. Since water profile data are extremely scarce, an attempt was made to formulate a general analytic expression for the vertical distribution of fog water as discussed in section III-3.
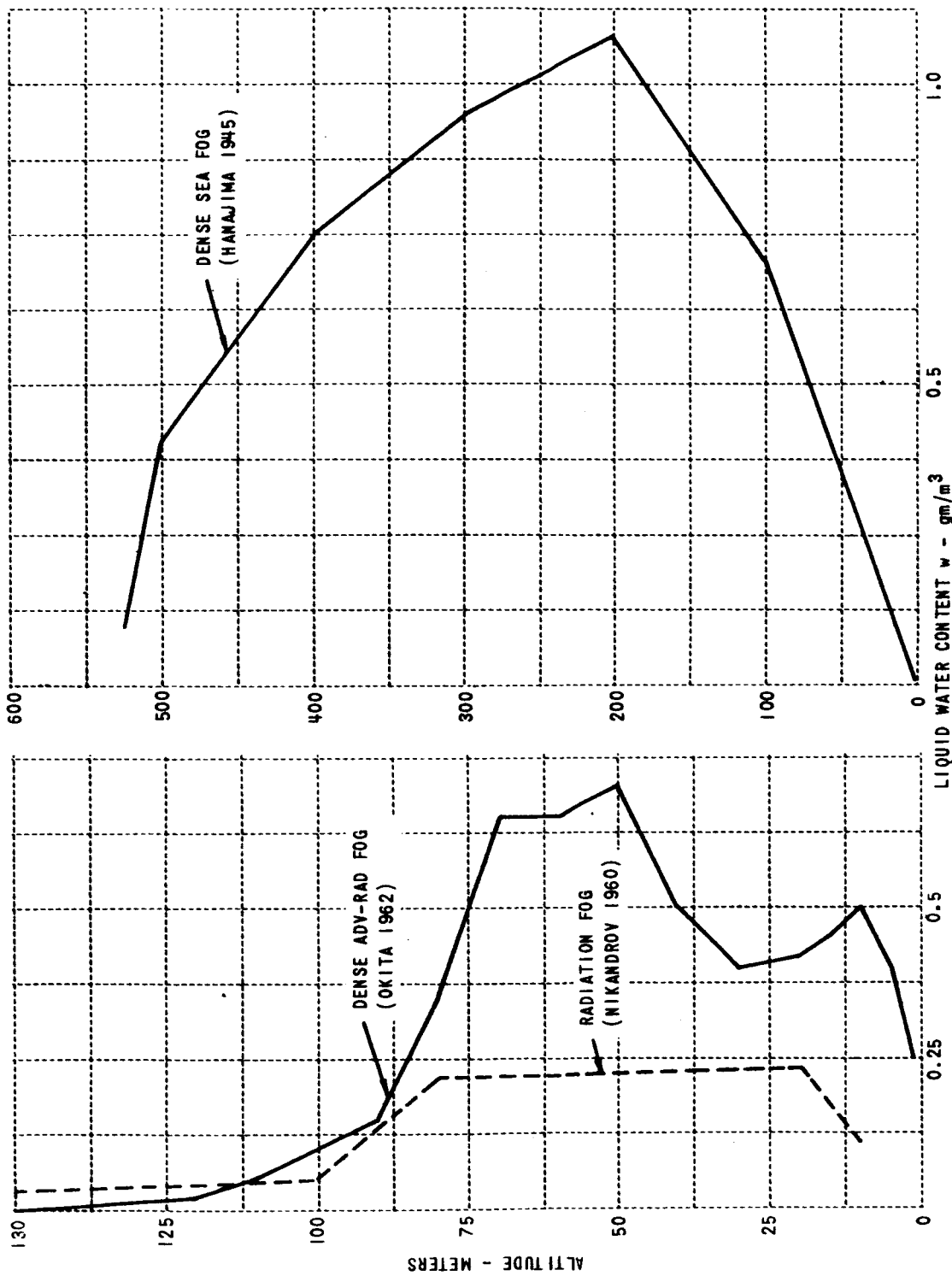
Figure I VERTICAL DISTRIBUTION OF LIQUID WATER CONTENT IN FOG

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Another important characteristic of fog is its drop size distribution. Based on experimental data from fog and clouds, Best (1951) found that drop size distributions can be represented by the formula

$$1 - F = exp(-x/a)^b \tag{1}$$

where F is the fraction of liquid water in the air consisting of drops of diameter less than x ; b is approximately 3.3; and a is related to liquid water content w by

$$w = 1.1 \times 10^{-3} a^{1.79} \tag{2}$$

Values of w were taken from the fog models (Table II) and expressions (1) and (2) tabulated. The results are plotted in Figure 2. These curves show the contributions which the different droplet sizes make to the total liquid water content of the fog. The half widths[*] of the functions in Figure 2b extend from 7 to 17 microns for inland fog and from 8.5 - 21 microns for coastal fog. Referring to Figure 2a, it can be seen that each of these droplet-size intervals encompasses approximately 80 per cent of the fog liquid water. Similarly, Figure 2c, d is for upper fog levels. The curves suggest that these drop sizes may be of major importance in the development of fog-modification concepts.

One instructive way to assess fog severity and its implications on modification attempts is to evaluate the total amount of condensed water in a vertical column through a given fog. It is convenient to consider a column with a 1 m^2 cross section. For complete fog dissipation, this columnar quantity of water multiplied by the lateral extent of clearing represents the total amount of water that must be either evaporated or precipitated. In practice complete

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[*]The width of the line measured between two points at which the function is half its peak value.

Figure 2a FRACTION OF FOG LIQUID WATER CONSISTING OF DROPS WITH DIAMETERS LESS THAN X

Figure 2b CONTRIBUTION BY DROPLETS OF VARIOUS SIZES TO FOG LIQUID WATER CONTENT (LOW LEVELS)

Figure 2C

Figure 2c FRACTION OF FOG LIQUID
WATER CONSISTING OF DROPS WITH
DIAMETERS LESS THAN X

Figure 2d CONTRIBUTION BY DROPLETS
OF VARIOUS SIZES TO FOG
LIQUID WATER CONTENT

(UPPER FOG LEVELS)

fog dissipation would not be necessary; it would be sufficient merely to improve the visibility within a fog to the point where aircraft landings and take-offs were possible. In this case only a fraction of the total column water need be evaporated or precipitated, or possibly shifted to a different size distribution favoring increased visual range. We will also consider the amount of water in the lowest 100 meters since clearing of fog to this altitude would enable aircraft instrument landings.[*]

Accordingly, we have calculated the total amount of liquid water in a column and the amount of liquid water in the lowest 100 meters for the three fogs indicated in Figure 1. The results are tabulated in Table III.

Table III Fog Water Content

		Inland Fog (Radiation)	Coastal Fog (Adv-Rad)	Coastal Fog (Sea Fog)
1.	Depth of fog	150 m	130 m	550 m
2.	Total liquid water in 1 m^2 column	20.8 g	46.2 g	390 g
3.	Total liquid water below 100 meters in 1 m^2 column	19.4 g	45 g	35 g
4.	Total liquid water in arbitrary runway "zone" (100 m high, 50 m wide, 2000 m long)	1940 kg	4500 kg	3500 kg

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[*]ILS (Instrument Landing System) landing limits for jet aircraft involve ceilings and visibilities of 300 feet and 0.75 mile respectively. GCA (Ground-Controlled Approach) minimums are less restrictive than the ILS limits.

These fog water values help not only to indicate the scope of the problem but also to enable calculations of energy requirements for suggested modification concepts. One revealing feature of the table is that the total water in the lowest 100 m of these markedly different fogs varied by only a factor of two -- this despite the fact the extremely dense sea fog contained about 19 times as much total condensed water as the typical inland fog. This feature of fogs, if representative, could prove an encouraging factor in fog modification.

3. Mathematical Fog Model

One of the objectives of the first year's research was to develop a mathematical model of fog that would describe the distributions of fog properties in time and space as functions of the imposed boundary conditions. A dynamic model is needed both to specify the distributions of temperature, humidity, and liquid water content, and also to provide insight into the mechanisms and energy exchanges acting in the formation and maintenance of naturally occurring fogs.

Three cooling mechanisms have been suggested and frequently called upon in the past to explain radiation and advection fogs: 1) radiational cooling of the earth's surface, leading to transport of heat from the air to the ground, 2) direct radiational cooling of moist air and already existing fog, and 3) mixing of moist air masses of different temperature. Over the years there have been numerous attempts to assess the relative importance of these three factors in the production of different types of fog. Most investigators have sought to demonstrate the predominance of one factor. For example, Emmons and Montgomery (1947) showed, with certain assumptions, that it was not possible to produce supersaturation by cooling an air mass from below. They therefore concluded, as have others, that fog was usually the result of radiational cooling of moist air. Taylor (1917) also decided that turbulent transfer of heat to a cold surface would not cause fog, but he explained observed advection fogs on the basis of mixing of warm and cold masses of moist air. Lyons et al. (1962) state that this mixing effect is so small as to be negligible and, therefore, that radiational cooling is the most important cause of fog. The apparent differences of opinion stem partly from the fact that different types of fogs were involved.

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In a recent paper, Rodhe (1962) comprehensively reviewed the subject of fog formation and suggested that two or more factors are usually important in any natural fog. Rodhe's conclusions point up the importance of the mixing process, but he also shows how radiational effects contribute. Like previous authors, Rodhe minimizes the importance of direct cooling of the air by molecular and eddy heat conduction, but his mathematical formulation makes it possible to retain this effect if it does contribute. Therefore, we have used his analysis as a starting point in our formulation of a dynamic fog model.

The vertical eddy flux of total heat H in the boundary layer will be

$$H = -\rho \left[c_p + T' \frac{d}{dT'} \left(\frac{LV'}{T'} \right) \right] K_H \frac{\partial T'}{\partial z} \tag{3}$$

and similarly, the vertical eddy flux of total water content E is

$$E = -\rho K_E \frac{\partial r}{\partial z} \tag{4}$$

In the equations, ρ is the density of the air, z is the vertical coordinate, L is the heat of vaporization, K_H and K_E are the eddy exchange coefficients for heat and water, respectively, T' is wet-bulb temperature, the term in brackets in (3) is the specific heat of moist air, and r is the total water mixing ratio equal to the sum of the saturation mixing ratio V' and the liquid water w

The vertical flux of water may be rewritten

$$E = -\rho K_E \frac{dr}{dT'} \frac{\partial T'}{\partial z} \tag{5}$$

where we consider the total differentials dr and dT' to be evaluated over vertical differences only; i.e. $dr = (\partial r/\partial z) dz, \quad dT' = (\partial T'/\partial z) dz.$

Now, eliminating $\partial T'/\partial Z$ between equations (3) and (5), we obtain a differential relation between water content and wet-bulb temperature:

$$\frac{dr}{dT'} = \frac{E}{H} \frac{K_H}{K_E} \left[c_p + T' \frac{d}{dT'} \left(\frac{LV'}{T'} \right) \right] \tag{6}$$

Equation (6) is given by Rodhe except that he assumes $K_H = K_E$. We retain the ratio K_H / K_E in order that the effect of different exchange coefficients can be taken into account.

The application of this fundamental result, equation (6), can be demonstrated with reference to a T', r diagram, similar to the temperature, vapor pressure diagram used by Taylor (1917) in his classic description of fog formation. In Figure 3, the curved solid line represents the relation between wet-bulb temperature (equal to dry bulb temperature at saturation) and saturation mixing ratio at a pressure of 1000 mb. The dashed line is one possible integral of equation (6), with E/H and K_H/K_E constant. Where the dashed curve lies to the left of the solid curve, the state is one of supersaturation, and fog would exist. The amount of liquid water at any temperature is given by $r - V'$ i.e. the distance between the saturation curve and the actual curve. The dashed line shown in the example might result from an advection fog situation. If air at some upper level, point B, is at a wet-bulb temperature of 11°C and relative humidity of 93%, and the corresponding surface conditions are -2.5°C and 91 percent, supersaturation will exist at the intermediate levels indicated. The figure shows that liquid water contents of up to 0.3 g m^{-3} could be realized in such a fog, recognizing that the assumed 11°C temperature difference might be uncommon. For two air masses represented by points C and D, it is evident that mixing could not produce supersaturation or fog.
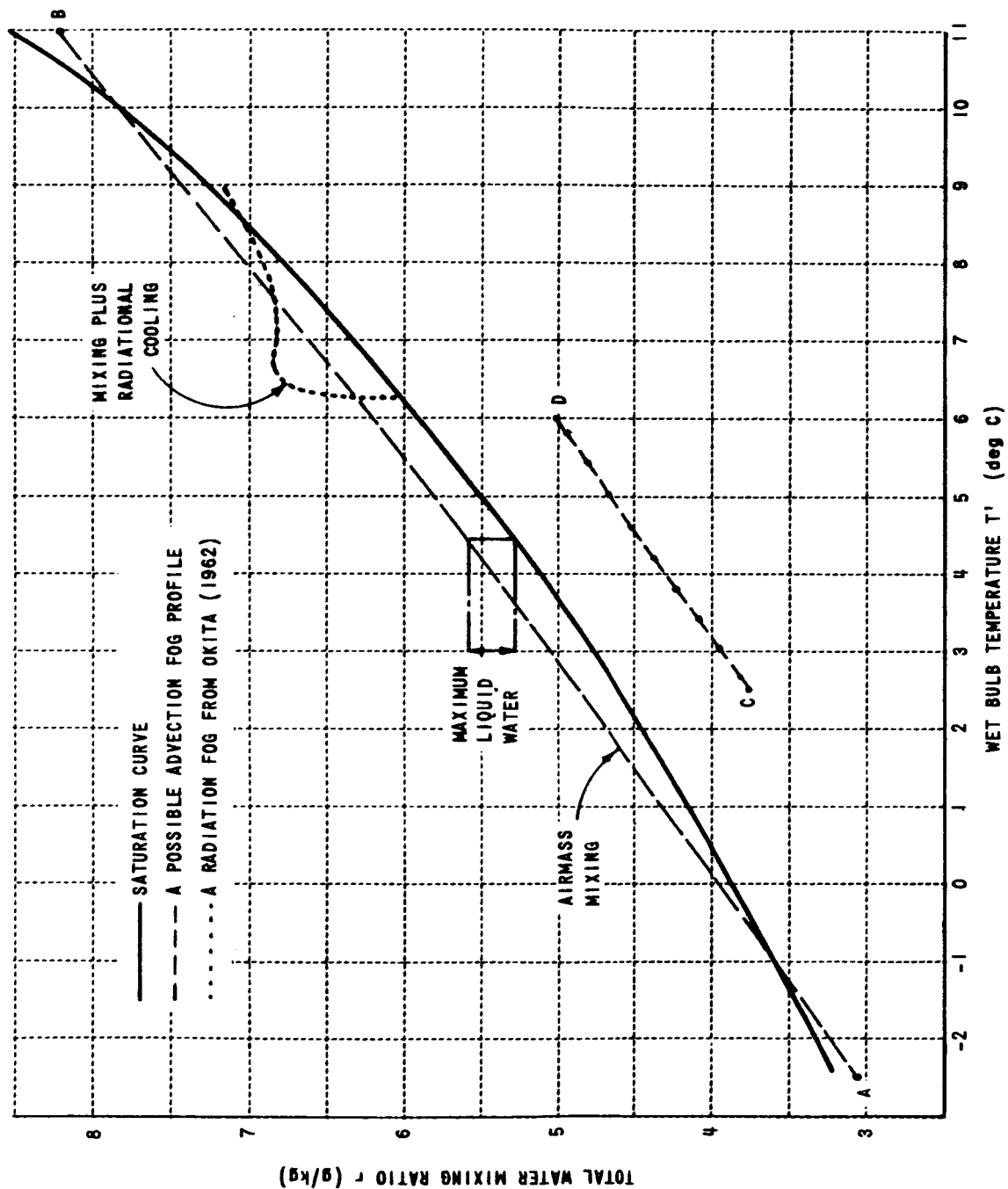
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Figure 3 PHASE DIAGRAM OF FOG FORMATION

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Two conclusions follow directly from the discussion thus far. First, in the simple case where the ratios E/H and K_H/K_E are constant (for these conditions the temperature and total water profiles are similar), fog can be formed in a moist surface boundary layer if the warmer air aloft is near saturation. Though moisture is withdrawn from the air by vertical eddy diffusion, supersaturation can occur at intermediate levels. Secondly, substantial vertical temperature gradients are needed to give appreciable liquid water. Thus, the pure mixing mechanism may be adequate to explain some advection fogs where the air mass is considerably warmer than the underlying surface, but the effect does not seem large enough to explain many observed radiation and advection fogs where vertical temperature gradients through the fog are weak.

To obtain higher liquid water content with small temperature differences, we must consider cases for which E/H is not constant with height. The most important effect which would lead to variation of E/H is radiational cooling of the air and fog. If one part of the boundary layer is losing energy by radiation, turbulent heat transport acts to balance the losses; thus H will vary in the vertical. There is no reason to expect E to change with height in this situation and as a result, E/H is no longer constant and the T', r curve may have an entirely different shape from that for constant E/H.

The previously mentioned advection-radiation fog documented by Okita (section III-2) enabled a curve of T' vs r to be plotted (dotted line in Figure 3). A crude calculation of the radiation required to produce such a curve was made and our results indicate that radiational cooling of the fog top was essential in the growth and maintenance of this fog. Hence, the ability to account for a variable E/H ratio is an important feature of a fog model.

Case studies of a few Buffalo fogs and of fog data reported by previous investigators have distinguished advection fogs, radiation fogs, and combinations of the two, all of which may be described by the above theory using appropriate boundary conditions. However, some observed cases have shown the necessity for considering additional factors.

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An advection fog that occurred in Buffalo, N. Y. on 14 May 1963 demonstrated the need for invoking two dimensional considerations in fog modeling. The fog was preceded by overcast skies which should have minimized the effects of radiational cooling; the wind speed exceeded 10 knots throughout the period of fog. The temperature differences between the surface and the fog top was less than $2^{\circ}C$ at the time of observation. A process of pure vertical mixing, operating within this temperature gradient, could lead to fog with a maximum liquid water content of 20 mg/m^3. When compared to typical advection fog water contents of 170 mg/m^3 (as per our structural fog model), it is clear that mixing alone could not have produced the dense fog observed unless the temperature difference at the point of initial fog formation was substantially greater than $2^{\circ}C$. There is evidence that the latter, in fact, was the case and that the fog formed over Lake Erie and then moved inland with the wind. Thus, a two-dimensional model is desired in this case to describe modification of the fog as it is advected horizontally.

Little consideration has been given to date to the role of hygroscopic condensation nuclei in the formation of fog. This is a factor which we feel may be important in the realistic modeling of metropolitan fogs and coastal (ocean) fogs. All fog models encountered thus far have assumed that fog forms only when the relative humidity reaches 100% and that the liquid water content of a fog is zero until saturation occurs. Clearly, hygroscopic nuclei initiate condensation at humidities substantially below 100%, and could serve as sinks to capture water vapor that might otherwise condense harmlessly on the colder ground surface.

The first year effort has contributed to the understanding of fog dynamics and indicated a useful form for models of the more simplified fog types. It is now necessary to extend the basic model and cast it in a form to handle variables that are given or can be estimated from routine meteorological observations. The following continuation of effort is recommended:

a. Continue the analysis of fog observations to define the relative importance of variables and processes in our models.

b. Investigate the magnitude of the radiative cooling of the surface air layers and fog top under typical conditions.

c. Consider two-dimensional models of advection fog.

d. Evaluate the role of condensation nuclei in the fog formation process.

4. Droplet Surface Properties

Over much of its life cycle, fog represents a system in equilibrium i.e. water droplets in equilibrium with vapor molecules. Considered in this program were droplet properties, both electrical and thermodynamic in nature, which when altered might bring about a condition to upset this equilibrium. The droplet properties considered were temperature, vapor pressure, evaporation (or condensation) coefficient,* accommodation coefficient,* charge, and surface tension. These are not to be thought of as independent properties since a change in one may directly or indirectly alter the other properties.

Table IV lists typical values of these droplet properties compatible with warm fog conditions. Also indicated in the table are illustrative means by which these droplet properties can be changed. Subsequent sections of the report will deal in some detail with the method and magnitude of droplet changes and their implications on fog control.

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*The evaporation (or condensation) coefficient of a liquid is by definition the ratio of the observed rate of evaporation (condensation) under given conditions to the "expected" rate based on the existing saturation vapor pressure. In short it is the probability that a molecule striking the air-liquid interface will be ejected (adsorbed). The accommodation coefficient represents the degree to which gas molecules that strike a bulk liquid come into thermal equilibrium with the liquid.

Table IV Droplet Properties

Droplet Property	Typical Value for Warm Fog Droplets	Illustrative means of Altering Droplet Property
1. Temperature	5 to 10° C	Heating of Air
2. Vapor pressure	8.7 to 12.3 mb	Hygroscopic Nuclei
3. Condensation Coefficient	0.04	Fatty alcohol monolayers
4. Accommodation Coefficient	1	
5. Growth rate (r = 10μ, R. H.= 101%)	5 x 10^{-9} g/min; 4μ/min	Fatty alcohol monolayers
6. Surface tension	75 dynes/cm	Ionic surfactants
7. Charge	0 to 50 electron charges (little data available)	Ion attachment

5. Laboratory Droplet Growth and Evaporation Experiments

It is now recognized that a population of cloud or fog droplets possesses remarkably stable sizes that show little tendency to change except by general growth or evaporation of the entire population. Available moisture is shared by all the drops (in varying degree depending on their hygroscopic nature), but the small drops increase in size more rapidly than the large drops. This type of slow growth, which tends toward an aerosol of uniformly sized particles, discourages additional growth by coalescence and precipitation of droplets. (A discussion of fog droplet growth mechanisms is given in section III-6C.)

If means could be found to treat a portion of the drop population so that these drops would not compete for available moisture, the remaining drops might then grow sufficiently large to fall out of the cloud, capturing a portion of the small droplets in the process. Even if complete precipitation did not occur, the shift in drop size distribution and droplet concentration might improve visibility within the cloud. Our analytical studies and the experimental results which follow suggest that surface-active monolayers may offer this capability if effective droplet-treatment techniques can be devised.

A. Apparatus

In order to examine the behavior of simulated fog drops in the laboratory, it was necessary to devise apparatus that would create and maintain slight supersaturations with respect to water of a few percent and under. A supersaturation of one percent is rarely exceeded in the free atmosphere. Two techniques, which involve the controlled diffusion of water vapor by means of thermal gradients and by means of chemical gradients, were considered appropriate. Both were originally designed to study ionization tracks left by cosmic ray particles; very recently the chemical method has been used in cloud physics to measure condensation nuclei concentrations (Twomey, 1959). We have now adapted these two related techniques, in what may be an original application, to the controlled growth of suspended water droplets.

The thermal cloud chamber relies on a temperature gradient to produce supersaturation (Langsdorf, 1936). As pictured in Figure 4, the chamber contains two water reservoirs at different temperature, t_1 and t_2. Water vapor is transported from the upper, warmer reservoir to the lower reservoir, with the ambient water vapor pressure e of a given air parcel being linearly related to temperature (straight line in Figure 5). Saturated conditions e_s are represented by the curved line so that at any point in the chamber the supersaturations s is given by:

$$s = (e - e_s)/e_s . \tag{7}$$

Maximum supersaturation occurs approximately at the level corresponding to $(t_2 - t_1)/2$ or approximately at the mid-altitude of the chamber.

This type of cloud chamber, unlike an expansion chamber, is able to maintain "constant" supersaturations and to operate more readily at low supersaturations representative of atmospheric conditions. The thermal gradient cloud chamber requires careful design and operation since heat loss through the walls can result in unpredictable supersaturations.
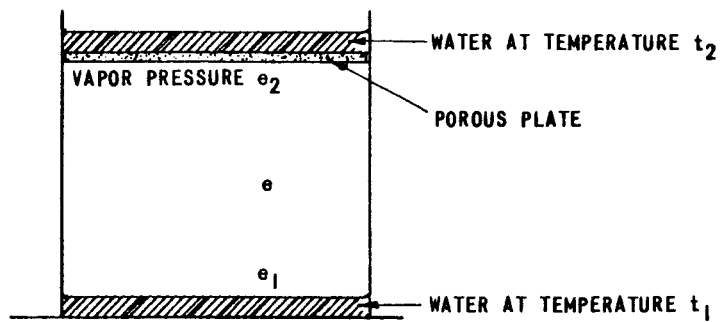
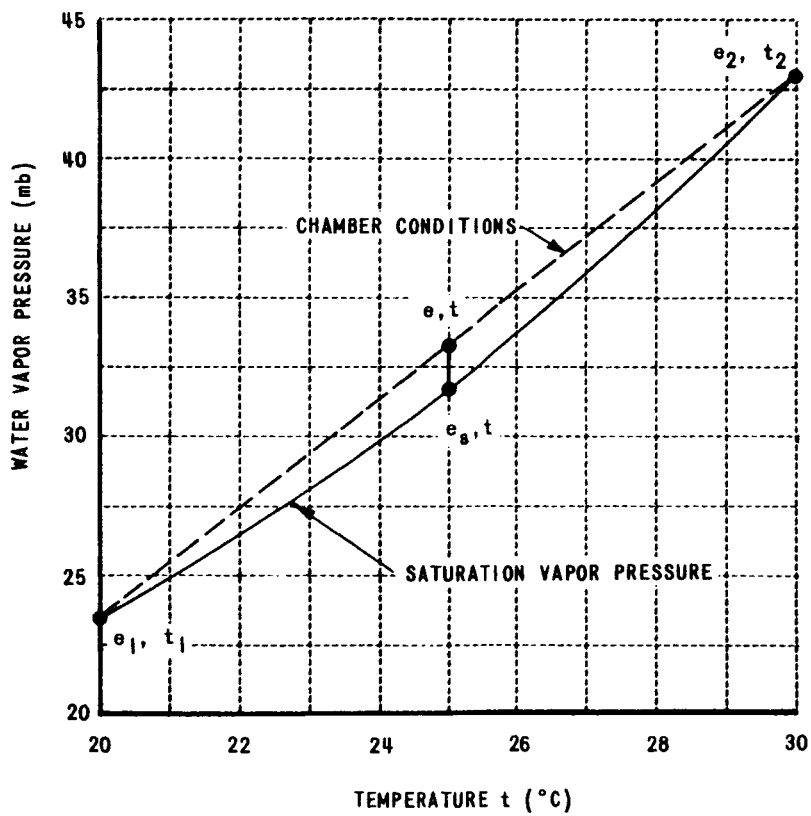Figure 4   THERMAL GRADIENT CLOUD CHAMBER

Figure 5   VAPOR PRESSURE IN A THERMAL GRADIENT
CLOUD CHAMBER

The chemical diffusion chamber of Vollrath (1936), described in some detail by Twomey (1959), overcomes the heat loss problem by operating at a constant temperature -- conveniently, at room temperature. The chamber is identical to the thermal gradient diffusion chamber except that the lower reservoir of water is replaced by an aqueous acid solution. Hydrochloric acid is customarily used since it is less surface active than most materials; the surface tension of aqueous HCl droplets differs by less than 1% from pure water. Of basic importance, the HCl-water system results in supersaturated conditions within the chamber. The maximum supersaturation, which occurs at a level 0.8 of the way from the acid to water reservoir, is a function only of the concentration of acid employed. For example, a 3% (by weight) HCl acid solution will produce a maximum supersaturation of 1%.

The water vapor pressure at the upper water surface is e_o , the saturation vapor pressure over pure water; over the HCl solution, the water and HCl vapors will have respective partial pressures of e_s and a_s . The values of e_s will be less than that of e_o in accordance with Raoult's Law for dilute solutions, i.e.

$$e_s/e_o = N/(N + n) = \text{mole fraction of the solvent} \qquad (8)$$

Thus, within the system water vapor will diffuse downward and acid vapor upward. Whether the transfer is by molecular diffusion or by turbulent mixing, the vapor gradients between reservoir surfaces will be linear so that the partial pressures of water and acid vapor (e, a) at any intermediate height can be readily calculated.

A droplet that forms in the HCl-water system will condense both moisture and HCl in constant ratio. The solution droplet will achieve equilibrium with some partial pressure of water vapor $e(a)$, which is a function of the acid vapor pressure at that point in the chamber. The departure of HCl solutions from the linear relationship of Raoult's Law are such that $e > e(a)$ and continuous droplet growth occurs. System supersaturation is given by

$$s = \frac{e - e(a)}{e(a)} \qquad (9)$$

Vapor pressure data for the binary system (HCl - H$_2$O) were obtained from the International Critical Tables (Vol. 3) and plotted in Figure 6. The resultant phase diagram was drawn for a 6% HCl solution to illustrate the foregoing critical variables.

We constructed a small chemical diffusion cloud chamber, as shown in Figure 7, that can with some modification also serve as a thermal gradient cloud chamber. Solution droplets and droplets treated with surface active agents were suspended in the chamber on glass, quartz, or spider-web filaments and comparative growth rates examined. Illustrative photomicrographs of three water droplets within the chamber are pictured in Figure 8.

To maintain a quasi-constant supersaturation, it is necessary to limit wall and filament condensation, which would otherwise deplete the system's vapor supply and prevent growth of the test droplets. In short it is essential that the flux of water and HCl vapor between reservoirs greatly exceed the depletion flux due to condensation. The vapor supply flux is increased by decreasing the distance between reservoirs. The reduction of extraneous condensation proved a painstaking task that was reasonably well achieved by:

1. scrupulous cleaning of the system to minimize condensation nuclei,

2. flushing of chamber air with bottled nitrogen, prior to a test, to remove nuclei, and

3. coating of the chamber walls and glass filaments with hydrophobic materials (e.g., paraffin, teflon, canada balsam). It was later found that the best results are achieved with a chamber made of lucite.

As will be observed in subsequent droplet-growth figures, chamber supersaturation can be maintained reasonably constant for periods of up to about 20 minutes. Thereafter, a gradual reduction in supersaturation due to increased wall condensation causes droplet growth rates to decrease. This type of competitive growth and supersaturation reduction corresponds closely with actual conditions in a cloud.
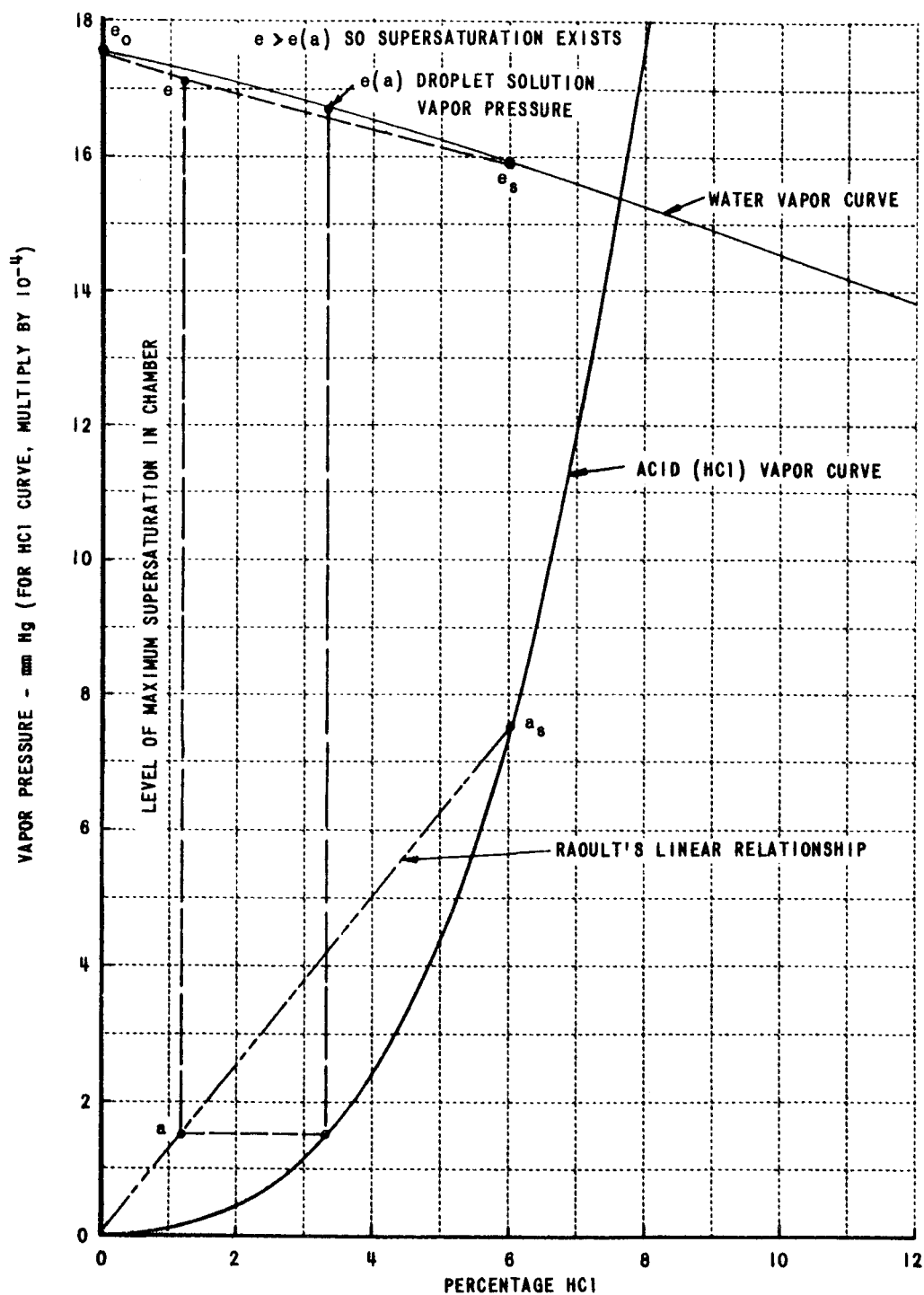
Figure 6 PHASE DIAGRAM FOR HCl–WATER SYSTEM --
CHEMICAL DIFFUSION CLOUD CHAMBER WITH 6%
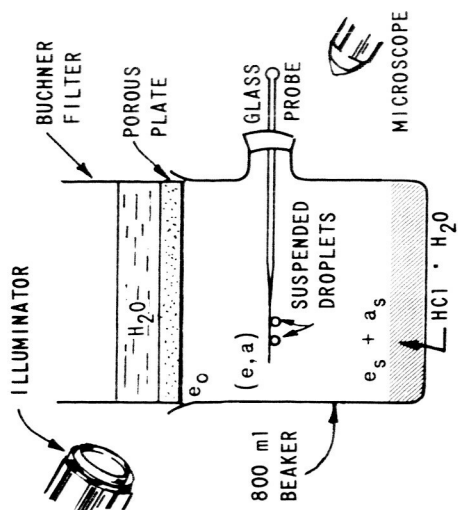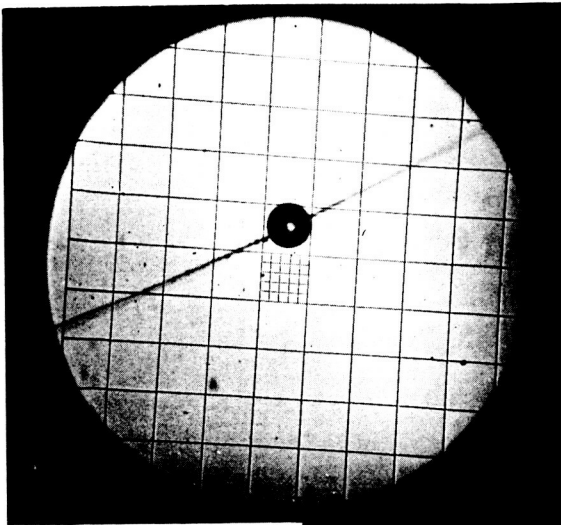ACID CONCENTRATION

26

Figure 7   CHEMICAL-GRADIENT DIFFUSION CLOUD CHAMBER

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SCALE: 67 MICRONS PER SMALL DIVISION
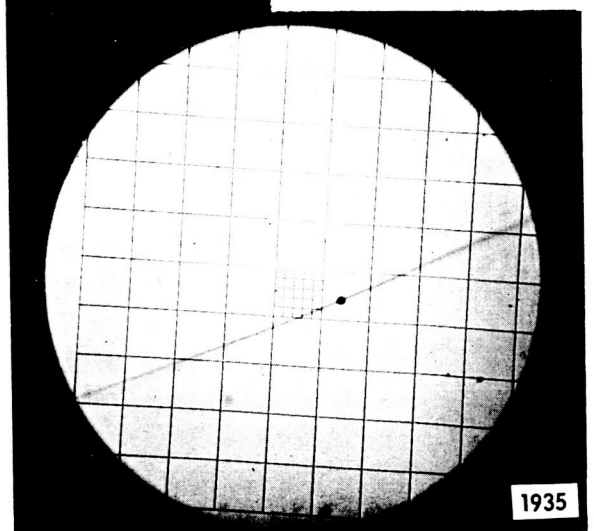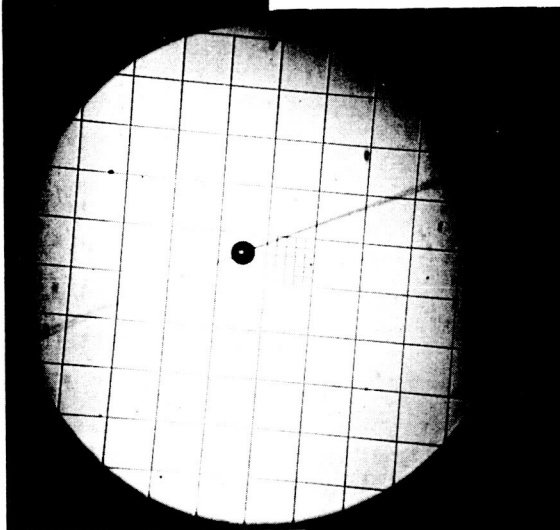335 MICRONS PER LARGE DIVISION

Figure 8 WATER DROPLETS SUSPENDED ON
GLASS FILAMENTS IN CLOUD CHAMBER

1935

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The chemical system can only be used with materials that do not react appreciably with aqueous HCl. Actually, fog droplets are frequently condensed on acid nuclei, although droplet acidity probably does not exceed 0.2% by weight. (Industrial combustion products, which include acid molecule complexes, commonly serve as condensation nuclei in inland fogs; chlorides and nitrates are effective nuclei in coastal fogs.)

Two pieces of fog-simulating apparatus were used to create large populations of droplets in the laboratory for experiments involving the study of droplet growth by coalescence. Fog was readily generated in a 200 liter cold box by cooling warm moist air to its dewpoint. The drops so produced, at temperatures of approximately $-2^{\circ}C$ to $+5^{\circ}C$, were mostly smaller than 15 microns diameter. Larger drop size distributions, from approximately 5 to 50 microns diameter, were obtained by mechanically atomizing water with a spinning disc. It is recognized that strong electrification effects are frequently associated with this method of producing droplets and that these effects must be accounted for.

B. Monomolecular Films on Droplets

Certain surface-active monolayers, such as the fatty-alcohols hexadecanol and octadecanol, are well known retardants of water evaporation when spread on ponds and reservoirs. Numerous investigators, including Langmuir and Schaefer (1943) and Barnes and LaMer (1962) have measured resultant evaporation rates; Bradley (1955) and Eisner et al. (1958) have also considered the effect of monolayers on evaporating droplets of water.

The results are in general agreement: in a subsaturated environment, the evaporation rate of bulk water and, to a greater extent, small droplets will be greatly retarded. For example, Eisner et al. (1958) measured the lifetime of 10μ diameter droplets at $20^{\circ}C$ and 80 percent relative humidity to be 0.64 second for pure water and 656 seconds for water treated with a monolayer of cetyl-stearyl alcohol. The evaporation reduction is accompanied by a marked decrease in the evaporation coefficient, a decrease in surface tension, and an alteration in electrical properties (polarization) of the droplet surface.

Eisner, Quince, and Slack (1960) have shown, using an equation based on one derived by Fuchs (1959), that droplet evaporation can be expressed by

$$\frac{dm}{dt} = \frac{4\pi MD}{RT}\left(fP_T - P_\theta\right)\frac{r}{1 + D/r\,\gamma\alpha} \tag{10}$$

where

$\frac{dm}{dt}$ is the rate of evaporation (g/sec) of a droplet of radius

M the molecular weight of the liquid

D the diffusion coefficient for the vapor into the air

f the relative humidity expressed as a fraction

P_T the vapor pressure of the liquid at the ambient absolute temperature

P_θ the vapor pressure of the liquid at the droplet temperature

R the universal gas constant

γ mobility of water vapor molecules equal to $\left(RT/2\pi M\right)^{1/2}$

α the coefficient of evaporation (or condensation)

The evaporation coefficient α for water is approximately 0.04. With the addition of a fatty alcohol monolayer, α is reduced to values between 10^{-4} and 10^{-5}, causing the evaporation rate to decrease approximately by a factor of 10^2 to 10^3.

This sizeable effect on evaporating drops prompted us to consider the effect of monolayers on droplets in a slightly supersaturated environment typical of clouds and fog. To our knowledge, this application has not been previously considered. According to expression (10), the growth rate of a monolayer-treated droplet will also be greatly retarded. This prediction is based on the assumption that the evaporation coefficient and condensation coefficient are equivalent; such is the case for plain water but not necessarily for droplets coated with monolayers. Hence, to test the theory and to evaluate the possible effect of treating fog drops with monolayers, a number of experiments were conducted.

The first series of experiments with individually treated droplets in the cloud diffusion chamber (described in section III - 5A) indicated that droplet growth rates could be markedly reduced. Typical results of these initial experiments are shown in Figure 9. Subsequent attempts to duplicate these results met with considerable difficulty. A careful step-by-step evaluation of possible variables involved led to ultimate reproducibility of results and a much better understanding of the behavior and limitations of monolayers. These findings will be summarized now and then followed by a general discussion of monolayer effects and details of the experiments.

1. The form of the droplet evaporation equation (expression 10) is valid for droplet growth under supersaturated conditions. In the laboratory, the condensation coefficient α may be decreased from a value of 0.04 to approximately 2.5×10^{-5} by the monolayers with a corresponding decrease in droplet growth rate.

2. The degree of inhibition of droplet growth obtained is strongly dependent upon (a) compression or spacing of molecules of the monolayer film, (b) type of monolayer employed and (c) the presence (absence) of impurities.

3. Our most consistent laboratory results were achieved with monolayers prepared from a mixture of hexadecanol and octadecanol crystal s. The mixture provides comparatively high values of resistance to droplet growth for only modest degrees of film compression.

4. In the case of evaporating drops, a film of low compression will become more compact and thus more effective as the droplet evaporates. For drops in a supersaturated environment, however, growing drops will cause a film of low compression to become even less effective. (Much of our initial experimental variability was caused by variations in film compression incurred in the treating and subsequent handling of drops.)

5. In droplet evaporation experiments, we were able to reduce α to values of 1.2×10^{-5} to 3×10^{-5} using monolayers formed with a mixture of hexadecanol and octadecanol powder. The most effective monolayer ($\alpha = 1.2 \times 10^{-5}$) would cause a 10 micron radius drop to last approximately 560 seconds versus 1 second for an untreated water drop at 20°C and 40% relative humidity.

31
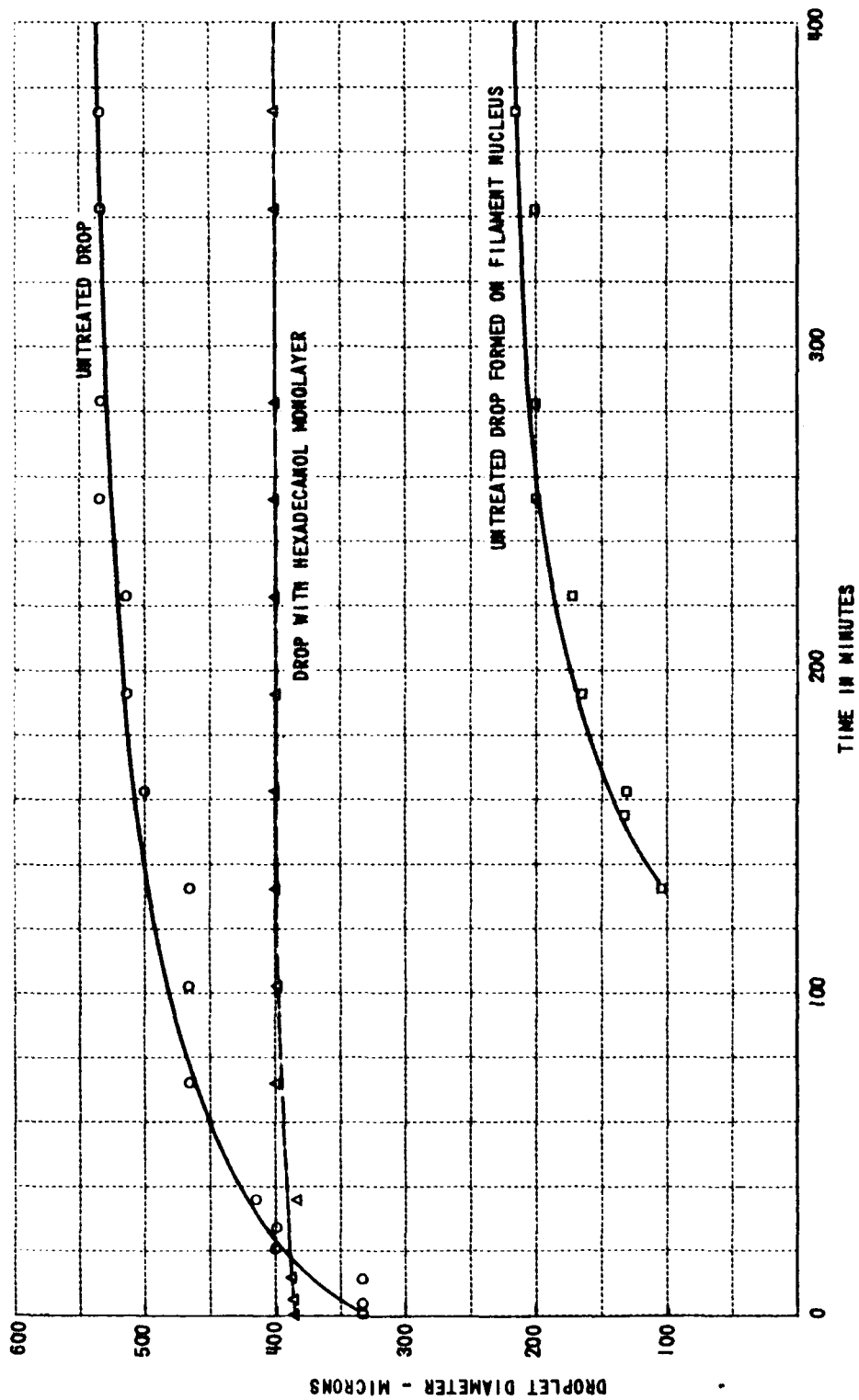
Figure 9 GROWTH OF DROPLETS IN CHEMICAL-GRADIENT DIFFUSION CHAMBER

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6. Impurities that become trapped within a monolayer can completely nullify the desired condensation retardation effect. Benzene, which formerly was used as a solvent for liquid dispersal of these fatty alcohols, constitutes such an impurity. Our data indicate that methyl alcohol is another deleterious solvent; petroleum ether is a satisfactory solvent. Salts and chemical compounds found in the atmosphere would undoubtedly vary the degree of effectiveness of monolayers.

The monolayer materials with which we experimented and some of their characteristics are listed in Table V.

Table V   Monolayer Properties

	Formula	Density g/ml	Melt. Pt.	Boil. Pt.	Solubility in water	ether
1. Hexadecanol (cetyl alcohol)	$CH_3(CH_2)_{15}OH$	0.817	49.3°C	190°C	i	s
2. Octadecanol (stearyl alcohol)	$CH_3(CH_2)_{16}CH_2OH$	0.812	59	210.5	i	s
3. Stearic Acid	$CH_3(CH_2)_{16}COOH$	0.847	69.4	283	.0348/100ml	s

It is known that these long chain molecules possess a hydrophilic component (hydroxyl OH ion or a carboxyl COOH ion) and a hydrophobic component (hydrocarbon). When in contact with water, the rod-like molecule aligns itself perpendicular to the surface so that the hydrophilic "head" seeks the water interface and the hydrophobic "tail" lies above. The hydrophobic "tails" of oriented molecules constitute the barrier to water vapor penetration. As Langmuir and Schaefer (1943) showed, such a single layer of material or monolayer is far more effective as a water barrier than multiple layers haphazardly oriented.

Figure 10, taken from Barnes and LaMer (1962), shows the dependence of water-evaporation resistance on surface pressure for monolayers spread on water reservoirs. Surface pressure, the customarily used measure of film compression, is a quantity with dimensions of force per unit length and is numerically equal to the difference between the surface tension of water and that of the film. Our laboratory experiments with droplets, rather than plane water surfaces, suggest that the resistance to droplet growth offered by mono-layers of hexadecanol and octadecanol follow similar trends to those indicated in Figure 10.
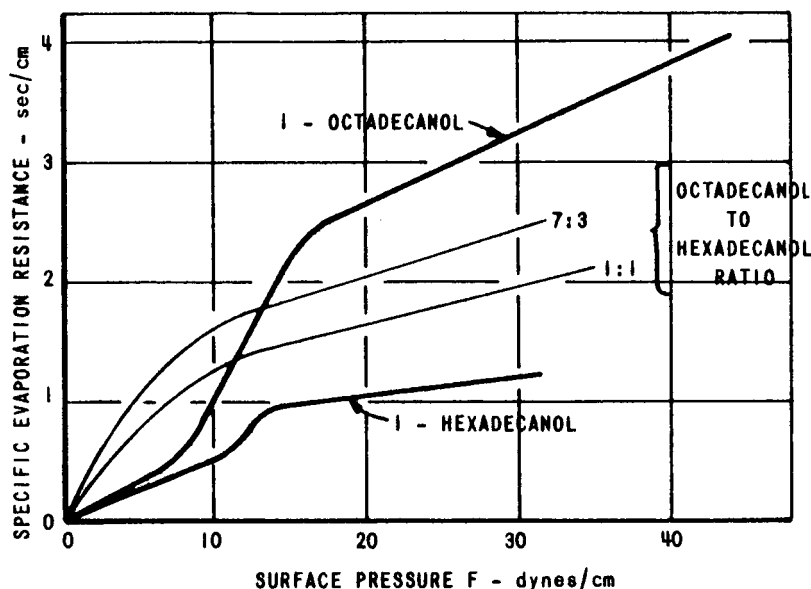
Figure 10   RESISTANCE TO EVAPORATION OF A PLANE WATER
SURFACE COATED WITH A MONOLAYER
(Barnes and LaMer, 1962)

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Three features of the curves are particularly noteworthy:

1. The specific evaporation resistance R (droplet growth resistance) of a fatty alcohol monolayer increases with compression of the film. This trend continues to a maximum surface pressure of about 50 dynes/cm, above which the film ruptures and R drops sharply. Though not shown, the R for stearic acid levels off with increasing surface pressure F.

2. Octadecanol, though more difficult to spread on water, is considerably more effective in inhibiting evaporation (condensation) than hexadecanol.

3. A mixture of octadecanol and hexadecanol is more effective than either single substance at low levels of film surface pressure ($F < 12$ dynes/cm). This is particularly important in the potential application to fog droplets where the achievement of large surface pressures corresponding to highly compressed monolayers may not be possible.

When dealing with monolayers on a flat surface, it is relatively simple to increase film pressures merely by mechanically compressing the film between movable walls or floats. With small droplets, this approach is not tenable. In these laboratory experiments, monolayer materials were prepared in powder and in liquid form and dispersed on water. A fine quartz probe was agitated in the bulk water solution and droplets that thereby collected on the probe were extracted along with some of the monolayer material. These drops were then transferred to a finer spider filament (2 microns diameter) mounted in the cloud chamber where growth rates were measured. It was discovered that film compression could be increased by causing partial droplet evaporation to take place. Figure 11 illustrates the increased effectiveness of a monolayer (hexadecanol plus octadecanol) as the film becomes more compressed through droplet evaporation. The solid curves represent the theoretical evaporation rate of water drops, uncorrected for ventilation effects.

Figure 11 DROPLET EVAPORATION AS INFLUENCED BY A COMPRESSING MONOLAYER
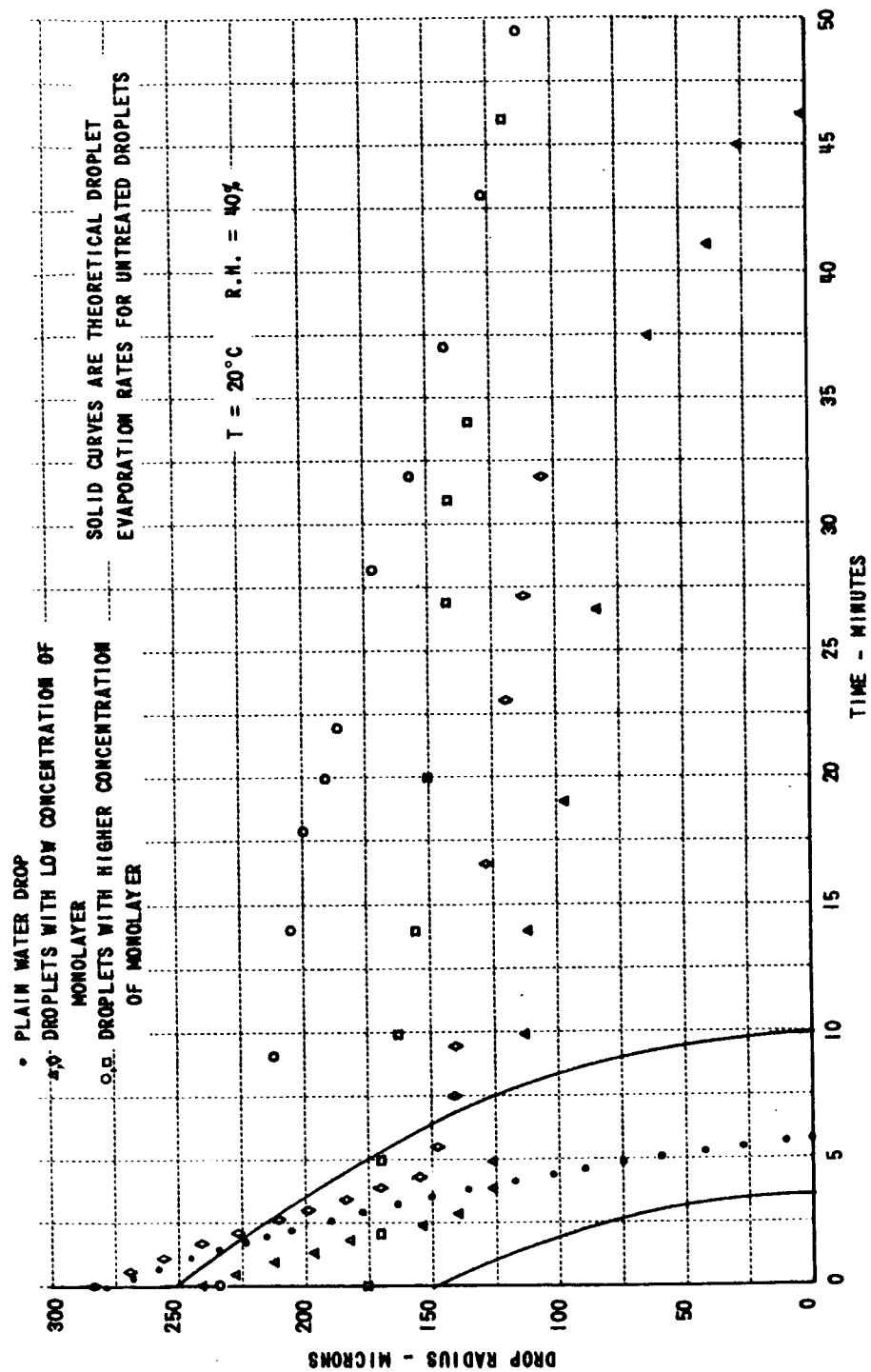
36

Film pressure could also be increased, within limits, by increasing the quantity of monolayer material dispersed on water; our measurements of surface tension, employing the ring method and drop-weight method, indicate that the maximum film pressure that can be achieved by increasing the amount of hexadecanol and/or octadecanol is approximately 30 to 35 dynes/cm.

It has been established that solution impurities, among them certain salts and benzene, constitute "holes" in a monolayer through which water vapor can readily penetrate. The resistance to water penetration of such impurities and of the film are analogous to parallel resistance in electrical circuits. Thus, a monolayer with "holes" equivalent to 1% of its surface area may have its water evaporation resistance reduced by approximately 90 to 99% (LaMer 1962). Frequently, impurities can be squeezed out of a monolayer by compressing the film, a laboratory option that does not presently appear transferable to fog applications.

Figure 12 represents one of our later typical experiments in which the foregoing variables were under control. It can be observed that the untreated droplet grew approximately in accordance with theory (solid curves) whereas no perceptible growth occurred with the monolayer coated drops. (It is of interest to note that after about 15 minutes in the cloud chamber, the growth rate of the untreated drop departed noticeably from the theoretical curves calculated on the basis of a constant supersaturation. Wall condensate acted as a vapor sink to gradually decrease the supersaturation, much as competing cloud drops cause supersaturation in a cloud or fog to approach zero.)

C.   Condensation Nuclei Deactivation

A related hypothesis for increasing the ultimate size of fog droplets is to deactivate a percentage of the condensation nuclei found in the free atmosphere prior to fog occurrence. The resultant fog might then consist of fewer, larger particles with less severe visibility characteristics. Hence, preliminary experiments were conducted to determine if hygroscopic condensation nuclei could be coated with fatty alcohol monolayers to prevent or retard condensation of water on the nuclei. Sodium chloride (NaCl) crystals, a common type of

37
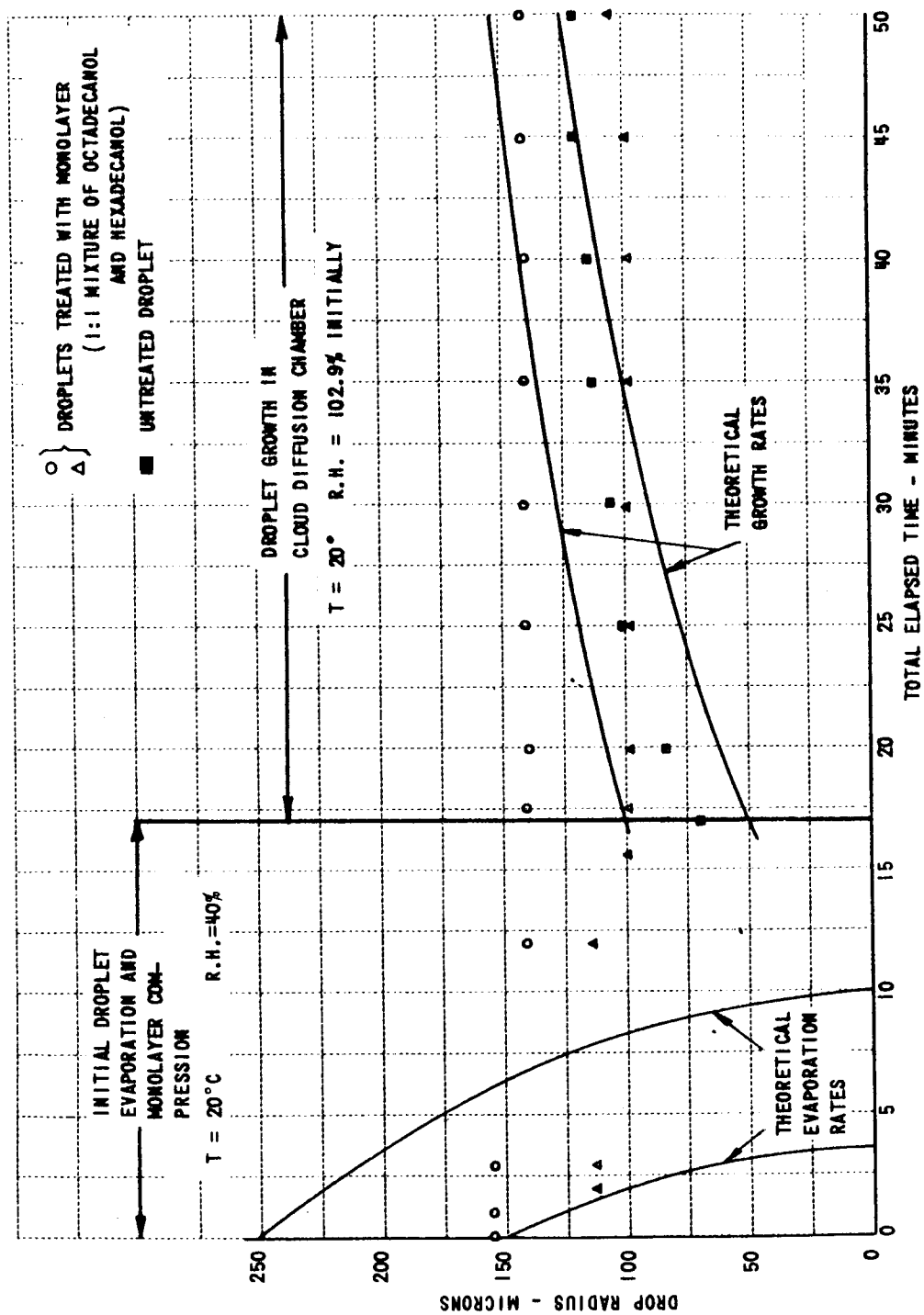
Figure 12 DROPLET GROWTH RETARDATION WITH MONOLAYERS

atmospheric condensation nucleus, were coated in various ways with a mixture of hexadecanol and octadecanol and their activity examined.

One coating method involved dissolving NaCl crystals in a liquid monolayer suspension and also in plain water. The salt crystals were recovered from the saline solutions by drying; drying time was substantially prolonged by the monolayer, thus attesting to the potential effectiveness of monolayers on NaCl solution droplets. After complete drying, the crystals were subjected to relative humidities of 90 to 100%. The rates of subsequent deliquescence of treated and untreated salt crystals were compared; no measurable difference could be detected.

Another coating method involved dusting of NaCl crystals with powdered octadecanol and hexadecanol. Some crystals were similarly coated with kaolin powder -- a non-hygroscopic inert clay material -- to simulate the simple physical barrier to diffusion that might be created by the alcohol powder. The subsequent rate of deliquescence of untreated NaCl crystals and of NaCl crystals powdered with kaolin were comparable. However, the deliquescence rate of NaCl crystals coated with octadecanol snd hexadecanol powder was retarded; total dissolution time of equal sized crystals was increased by a factor of three. These initial results suggest that further efforts to inhibit condensation nuclei are warranted.

D. Ionic Surfactants

Surfactant molecules are of two types, ionic and non-ionic. Ionic surfactants have some of the characteristics of salts - they separate into two oppositely charged ions in water. The "counter ion," consisting of a charged atom or small group of atoms, distributes approximately uniformly throughout the water. The major part of the molecule tends to concentrate at an interface, producing an ionized layer. A cationic surfactant yields a positive surface active ion and produces a positive charge layer at a surface. An example would be Benzalkonium chloride, where the counter-ion is Cl^- . Conversely, an anionic agent yields a negative surface active ion. The soaps, such as sodium stearate are good examples. In this case Na^+ is the counter ion.

Numerous investigators have suggested that the collision rate and coalescence[*] of droplets might be increased by selective surface charging of the droplets. Elton (1953) has postulated the use of ionic surface active agents to alter droplet surface charge in fog. Similarly, it is conceivable that the diffusional growth rate of water droplets will be influenced by ionic surfactants (Benton, et al., 1958). Lieberman (1960) has reported on laboratory experiments with ionic surfactants in which treated fogs became more, rather than less, persistent; recent Air Force field tests in Arcata, California with surfactants have indicated some promise (private communication with Bernard Silverman, Air Force Cambridge Research Lab.).

In order to assess both the collection efficiency and diffusional growth (evaporation) rates of droplets treated with ionic fluids, a number of surfactants were tested:

Anionic	Cationic	Non-Ionic
Sodium Heptadecyl Sulfate	Amine 220	3 types of nonyl
Sodium Tetradecyl Sulfate		phenyl poly-ethylene
Sodium 2-Ethylhexyl Sulfate		glycol ether

We measured the diffusional growth rate of individual droplets tested with the above surfactants in our cloud diffusion chamber. Figure 13 depicts two such experimental runs. As shown, no significant difference from the growth rate of untreated water droplets was observed.

Figure 14 depicts experimental results of evaporation of individual droplets treated with surfactants. Again, no significant difference from the growth rate of untreated drops was observed.

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[*] The term coalescence is used herein to denote the merging of two droplets that collide. For droplets in the range of a few microns diameter to a few hundred microns diameter, there are good indications(Fletcher, 1962) that coalescence always occurs. Hence, increased coalescence with ionic surfactants may be meaningless for fog droplets.

SOLID CURVES ARE THEORETICAL DROPLET GROWTH RATES FOR
UNTREATED DROPLETS

O "PLAIN" WATER DROPLET
△ DROPLET COATED WITH CATIONIC SURFACTANT
☐ DROPLET COATED WITH ANIONIC SURFACTANT
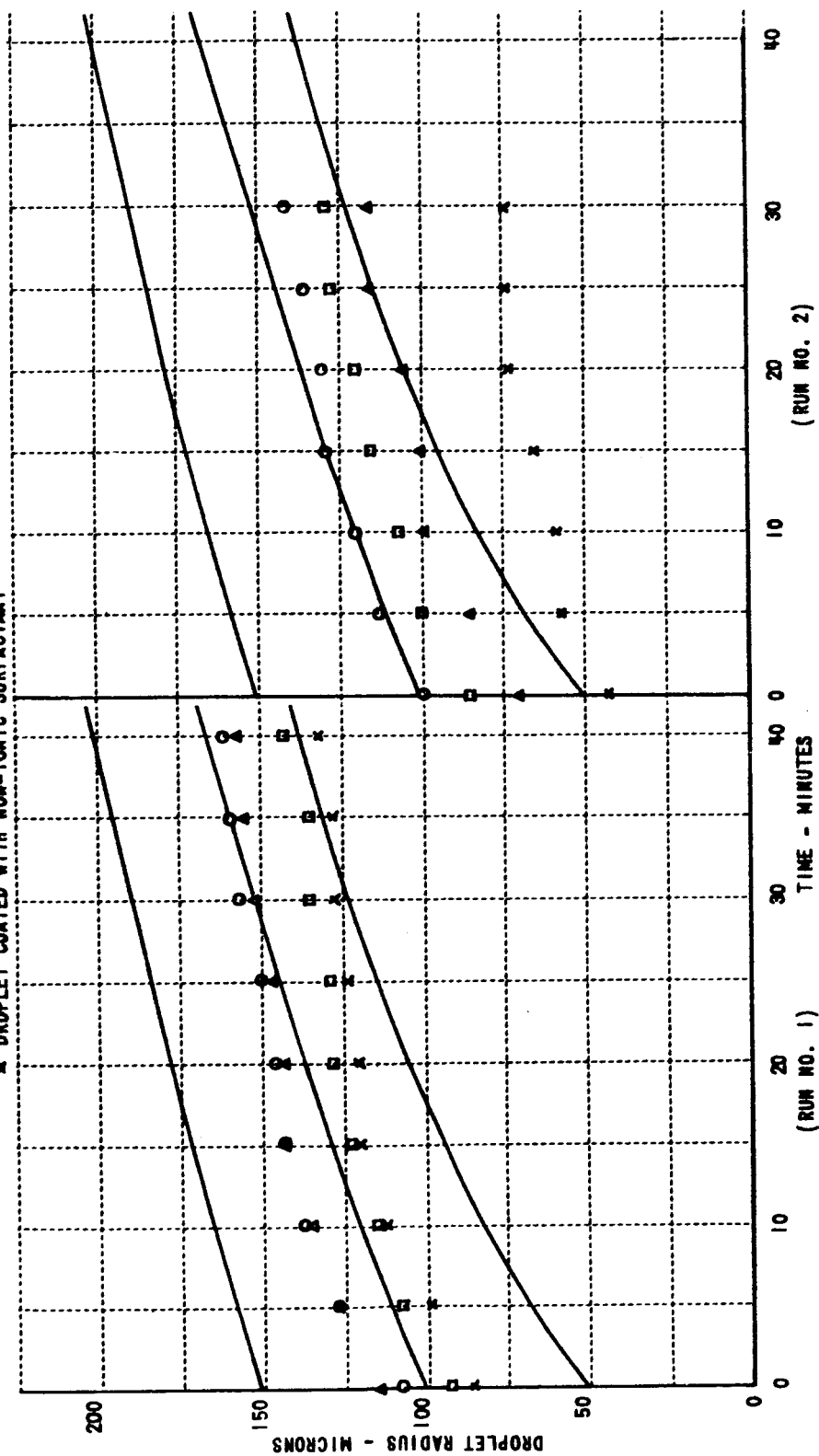✕ DROPLET COATED WITH NON-IONIC SURFACTANT

Figure 13 GROWTH OF DROPLETS TREATED WITH IONIC SURFACTANTS IN A CHEMICAL
DIFFUSION CLOUD CHAMBER -- SUPERSATURATION = 2.9%

41
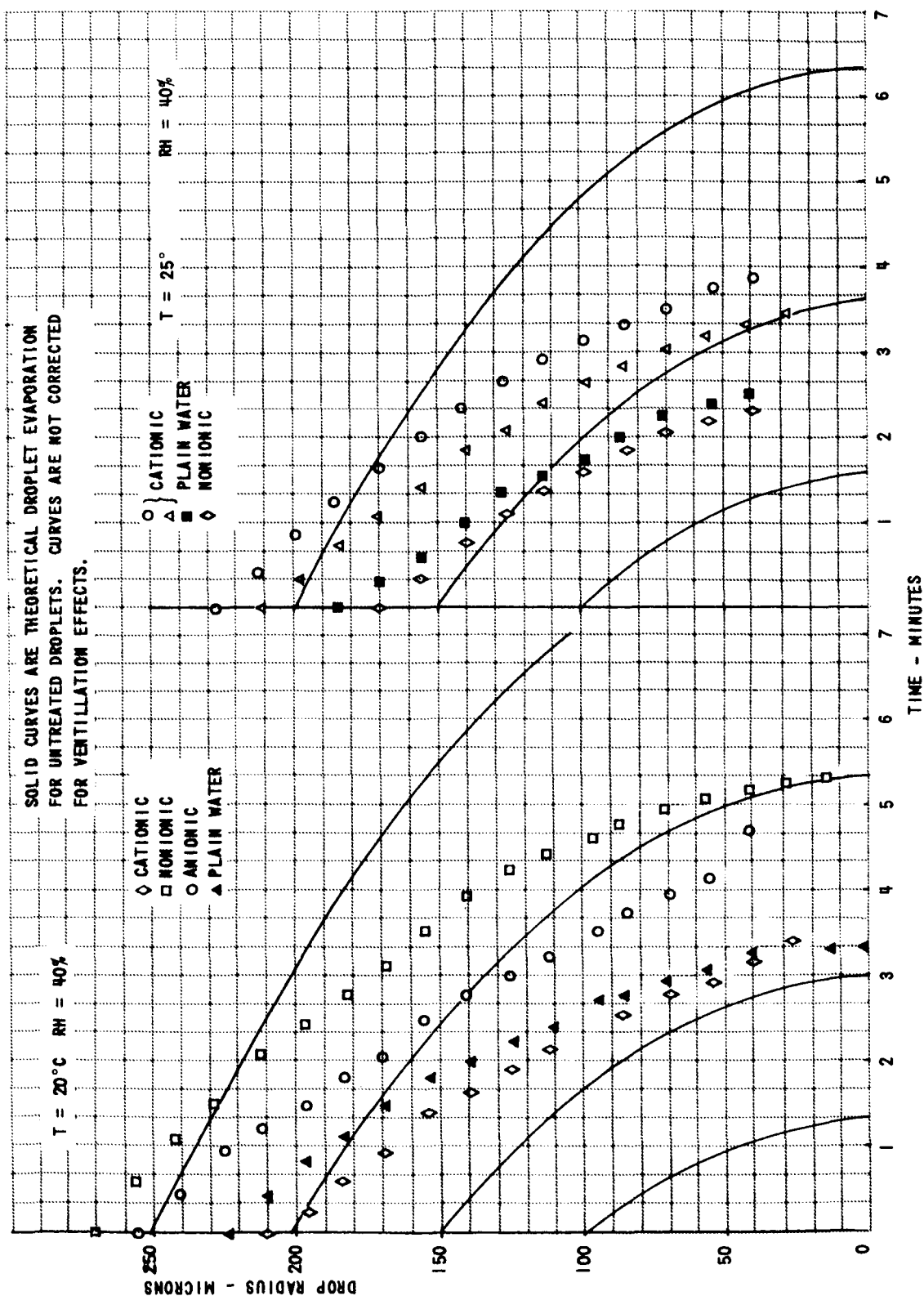
Figure 14 EVAPORATION OF DROPLETS TREATED WITH IONIC SURFACTANTS

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An attempt was made to assess the collection efficiency of droplets treated with ionic surfactants. Treated droplets of each type plus plain water drops were suspended on a spiderweb filament in a stream of "cloud" droplets (5-50 microns diameter) produced by a mechanical atomizer. When the treated droplets were subjected to the moving cloud (velocity approximately 1 m/sec in the region of the test droplets), they oscillated along the filament a distance of two to three droplet diameters. In moving along the filament, these droplets collected cloud droplets that had impinged on the filament. The abnormally high growth rates thereby obtained negated the experiments. Some tests were also made wherein the test droplets were suspended in a slowly moving cloud (drops less than 15 microns diameter) created in the cold box. Collection efficiency of all the suspended drops, including the plain water drops, were comparable but minimal i.e. close to the minimum sensitivity of the observing system. As a result of the experimental difficulties encountered, no conclusive results can be presented at this time.

6. **Fog Alteration Concepts**

A. **Program Findings**

The monolayer results discussed in section III-5B suggested three possible fog alteration concepts. Frontal fogs are caused by precipitation falling into a colder air mass, whereupon partial evaporation of the raindrops increases the ambient dewpoint until fog is formed. Stabilization of the rain drops with monolayers would retard evaporation and conceivably inhibit fog formation. The sizeable task of effectively treating the rain drops or perhaps the cloud prior to precipitating is as yet an undetermined factor.

Secondly, as referred to earlier, monolayer treatment of a portion of the drops in air mass fogs might cause selective growth of the remaining drops to the precipitation stage. However, our laboratory experiments indicated that to stabilize growing droplets in a supersaturated environment, one must begin with a compressed monolayer (unlike the case for evaporating drops where an initial inadequate film will automatically become compressed). We are not presently optimistic about being able to accomplish this in the free atmosphere.

Thirdly, preliminary experiments suggested that the activity of condensation nuclei might be decreased with monolayers. In this way subsequent haze and fog conditions might be characterized by fewer, larger particles and better visibility.

It is our growing contention that practical development of any of the above concepts will necessitate dispersal of monolayer materials in gaseous form. Whether this can be done without altering the desired properties of the monolayer is presently under study. Dispersal of monolayers in liquid or powder form appear impractical in terms of obtaining a high enough collision efficiency between monolayer particles and fog drops.

The diffusional growth and evaporation experiments discussed in section III-5D enable one to lay to rest thoughts of altering with ionic surfactants droplet growth (or evaporation) by diffusion. While these surfactants alter a droplet's surface tension (60% reduction) and surface ion distribution, they have no apparent effect on droplet vapor pressure and condensation coefficient. These latter variables affect diffusional droplet growth as shown in equation 10 of section III-5B.

That ionic surfactants can significantly alter coalescence rates of droplets remains to be verified. Our initial experiments to examine this possibility were inconclusive and will be repeated with more suitable apparatus.

B. Review of Fog Modification Efforts

A brief review of fog modification efforts was made to determine if marginal concepts of the past might not be more feasible in terms of recent technology advances. Appendix B includes a partial listing of the numerous fog modification concepts that have been postulated.

In general we feel that time has not significantly enhanced the appeal of these techniques; in fact, certain concepts may be viewed with less favor. One possible exception is the Fido-type method of evaporating fog through the combustion of large quantities of fuel. This method was effective when used in the past and conceivably could be more effective today with more efficient burners and fuels of higher heat-to-water vapor output ratios. The general objections of operating expense, extensive equipment installation, and fire hazard would probably still prevail.

Further exploration of chemical methods of altering droplet properties, as has occupied much of our first year's effort, and droplet electrification concepts (see next section) warrant continued study.

C.   Electrification Principles

Cloud droplets can grow in one of two ways, by diffusion of water vapor and by collision (coalescence assumed) with neighboring drops. Neither process is very effective in fog. Diffusional growth proceeds very slowly owing to the large number of drops that compete for excess water vapor and thereby cause the slight fog supersaturation to approach zero. As an upper limit, for example, if a supersaturation of 0.1% could be maintained -- which is not likely -- it would take approximately one hour for a drop to grow to 50 microns diameter.

Natural collision of fog droplets as a growth mechanism is practically negligible. Theoretically, collisions can take place by random thermal agitation (Brownian motion) and by aerodynamic interactions of particles with differing terminal velocities. The former mechanism can be dispensed with making use of an extension of Smoluchowski's well established theory of particle agglomeration (Fletcher, 1962):

$$\frac{dn}{dt} \approx -10^{-6} n^2 \left(1 + \frac{10^{-5}}{r}\right) \tag{11}$$

where $\frac{dn}{dt}$ is the number of collisions per cm^3 per hour

n is the number of particles per cm^3 $\left[200 \text{ cm}^{-3} \right]$

r is particle radius in cm $\left[5 \times 10^{-4} \text{ cm} \right]$

The brackets contain representative values for radiation fog as given by our structural model (section III-2). The collision rate dn/dt for such an assumed fog of uniform droplets would be $.04$ cm^{-3} hr^{-1} or 40 liter^{-1} hr^{-1} -- clearly an insignificant rate for growth purposes.

Considerable work has been done on computing the collision rate (or efficiency) of different sized droplets possessing differing fall velocities. Most appropriate to drop sizes characteristic of fogs, Hocking (1959) determined the collection efficiencies of droplets r_2 , up to 30 microns radius, moving through an emsemble of smaller droplets r_1 (See Figure 15a). Collection efficiency E is defined as the fraction of those smaller drops lying within the volume swept out by the larger drop that will be collected. The curves in Figure 15a indicate, among other things, that:

1. E is zero for collector drops of 18 microns radius or smaller
2. As drop sizes become uniform, E approaches zero.
3. Drops of 20 microns radius, for example, will collect drops of 14 microns radius with an efficiency of about $.25$.

Hence, it is apparent that fog droplets, particularly those constituting radiation fog (typical size range of 5-35 microns diameter), will normally experience little growth by aerodynamic collisions. However, if the droplets possess appropriate charge, the collision rate may increase appreciably. Some collection efficiency calculations of Cochet (1951), based on a collector drop bearing 8×10^5 electronic charges falling through a cloud of smaller neutral droplets, are shown in Figure 15b. Note that a 20 micron drop, which normally will not collide with 10 micron drops, is expected to do so with an efficiency of 100 (!) when so charged. Indications of this sort prompted us to explore (1) the magnitude of charge normally found on cloud and fog drops and (2) electrification means whereby aerosol droplets can be made mutually attractive.
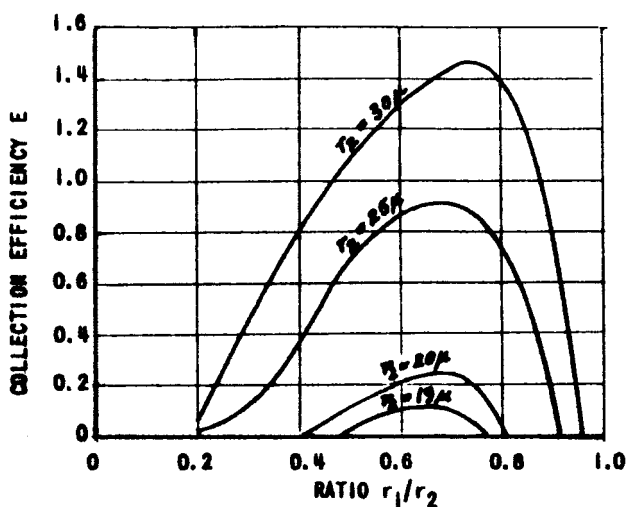
Figure 15a THE COLLISION EFFICIENCY OF SMALL DROPS. A DROP RADIUS $r_2\mu$ COLLIDES WITH DROP-
LETS OF RADIUS $r_1\mu$ LYING IN A HORIZONTAL CIRCLE OF AREA E TIMES THE CROSS-SECTION
OF THE DROP. THE VALUES OF E ARE SHOWN FOR VARIOUS DROP RADII AS FUNCTIONS OF
THE RATIO r_1/r_2 (Hocking, 1959).
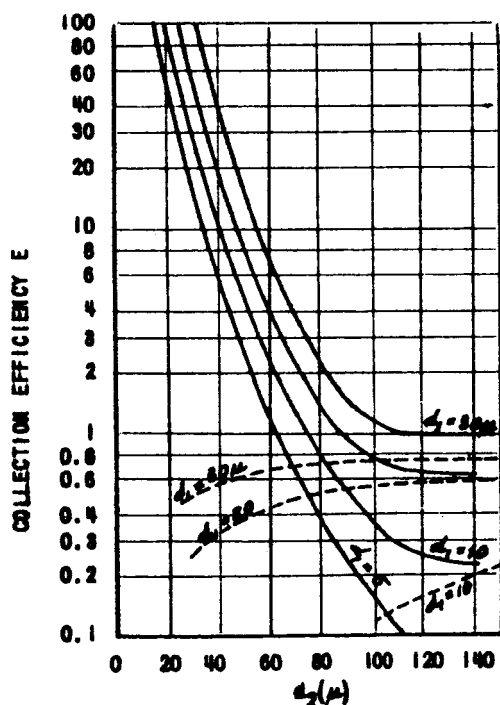
Figure 15b COLLECTION EFFICIENCY FOR A DROP DIAMETER d_2, BEARING A CHARGE OF 4×10^{-4} e.s.u.,
FALLING THROUGH A CLOUD OF DROPLETS OF DIAMETER d_1. BROKEN CURVES ARE LANGMUIR'S
CALCULATIONS FOR UNCHARGED DROPS (Cochet, 1951).

Above figures are from Fletcher (1962)

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Measurements of the charge carried by cloud and fog droplets are scarce and frequently relate to total charge measurements over a volume. Available droplet charge data are listed in Table VI, from which it can tentatively be inferred that the charge on cloud droplets (excluding thunderstorm situations) generally does not exceed a few hundred electron charges, often not exceeding a few tens of electron charges.

Table VI Charge on Cloud Droplets

Investigator	Volume (V) vs. Individual Drop (I) Measurement	Sample	Droplet Charge[*]
1. Reeder (1951)	I	.fog	50% drops < 200 ec
2. Gunn (1952)	V	a. clouds b. fog	a. + 32 ec/drop b. -30 ec/drop
3. Webb and Gunn (1955)	V	35 clouds	< 1 ec/drop
4. Twomey (1956)	I	clouds	20μ drops: 1000 ec
5. Philips and Kinzer (1958)	I	Mt. clouds	8μ drops: ±6 ec
6. Pudovkina and Katsyka (1960)	V, I	Mt. clouds	10-44 ave. ec/drop

Theoretically droplet interactions can be influenced electrically in the following ways:

1. Droplets bearing unlike charges will be attractive.

2. Highly charged drops (or solid particles) will polarize neutral neighboring droplets and attract them. Cochet's calculations are based on this mechanism.

3. Strong external electric fields will polarize neutral droplets and set up attractive forces between neighboring drops.

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[*]In practically all cases, numerous neutral drops were observed.

Though all of these mechanisms have been considered in the past for cloud modification, further exploitation of items 1. and 2. appears warranted. The foregoing brief analysis was conducted preparatory to engaging in forthcoming fog electrification experiments in the laboratory.

7. Supercooled Fog Dispersal

In 1946 Schaefer discovered that the stability of supercooled clouds could be upset by using dry ice to introduce ice crystals into such clouds. Vonnegut (1947) gave added impetus to weather modification efforts by showing that ice-forming nuclei such as silver iodide could serve the same purpose, though at slightly colder temperatures. Since the late forties, numerous investigators (including aufm Kampe, et al., 1957; Downie and Silverman, 1958; and Jiusto and Rogers, 1960) have demonstrated that supercooled fog and stratus clouds can be dissipated.

The occurrence of supercooled fog in this country is restricted to northern latitudes and to the winter season. A brief survey was made to determine the geographic regions where the frequency of supercooled fog is a maximum. Whether operational seeding programs should be considered for the affected regions is a matter dependent on such factors as fog frequency, air traffic density, responsible agency, seeding expense, commercial losses in rerouting, local civic interests, etc; it is not our intent here to engage in such deliberations.

Limited climatological data on supercooled fog frequency and combined data on total (warm and supercooled) fog frequency and temperature patterns for given areas were analyzed. Our findings can be summarized as follows:

a. The general region of maximum supercooled fog frequency in this country is in the Pacific Northwest. This region is delineated in Byers (1959) fog classification system as region 5 and includes portions of the states of Washington, Oregon, Idaho, Nevada and Utah.

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b. The wintertime fogs in the Pacific Northwest are principally of the high-inversion, radiation type. These fogs are characterized by their persistence, occasionally lasting for several days. Valley stations are particularly susceptible.

c. United Air Lines has tabulated available data, some of which are included in Appendix C, on supercooled fog in the Pacific Northwest. Their survey reveals six airports (see Appendix C) where 46 or more hours of super-cooled fog are recorded annually and at least three other airports where high fog frequencies are suspected but corroborative weather data are not available. Medford, Oregon tops the list with an average 213 hours of supercooled fog annually. December and January are the most critical months. During December 1959, Medford experienced an unusual 237 hours of supercooled fog with 36% (66) of the United flights being cancelled.

d. Over 50% of the supercooled fogs reported above occurred with surface temperatures between $29^{\circ}F$ and $32^{\circ}F$. Dry ice is the only known seeding material that is generally effective in this temperature range. Eadie and Mee's (1963) laboratory experiments suggest, furthermore, that even dry ice may not be effective at these temperatures unless the contact time between dry ice pellets and ambient air is several milliseconds. Hence, dry ice pellets falling at terminal velocity may be inappropriate. To overcome this limitation and to avoid the expense of aircraft seeding operations, we have demonstrated and advocated in the past the use of mobile tethered balloons with spaced packages of dry ice attached to the balloon line (Jiusto, 1963; Jiusto and Rogers, 1960).

e. Supercooled fog occurrence in other parts of the country is generally agreed to be of relatively minor concern. John F. Kennedy (Idlewild) Airport in New York, for example, averages about 12 hours of supercooled fog per winter (November through March) and about 330 hours of warm fog over the like five month period.

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V. APPENDICES

Appendix A - Fog Types and Classification - After Willet (1928) and Byers (1959)
(Examples of Regions of Occurrence in Parentheses)

A. AIR MASS FOGS

1. Advection

a. Land- and sea-breeze fog: warm, moist, land air in passing over cold water is cooled to the dewpoint. The fog is transported over land by afternoon sea breezes. (Summer phenomenon along New England coast.)

b. Sea Fog: like a. except that it arises from the cooling of sea air over a cold ocean current or cold coastal current. (Outer California coast in summer.)

c. Tropical-air Fog: caused by gradual cooling of tropical air as it moves poleward over the ocean or over land. (Most common open-sea fog type; causes widespread fog in the southeastern U. S. and east coast of the U. S.)

d. Steam Fog: advection fog caused by the passage of cold air over warmer water. This fog, which is strictly an over-water phenomena, is called "arctic sea smoke" in polar latitudes.

2. Radiation: nearly all fogs over land are wholly or partially due to radiational cooling of lower-level moist air.

a. Ground Fog: simplest type involving light winds, clear skies, and a surface temperature inversion. Its duration is confined to a single night. (Appalachian valleys)

b. High Inversion Fog: a winter time, land phenomenon resulting from more than a single night of cooling. The inversion extends through a deeper layer 100 to 600 m above ground often with a lower isothermal layer. (Low valleys of the far west in Winter.)

3. Advection-Radiation Fog: this fog is caused by nighttime radiational cooling of air that has moved inland from the sea during the day. (Great Lakes during Fall, California Coast.)

4. <u>Upslope Fog</u>: involves cooling of air in its dewpoint by adiabatic expansion as the air moves to higher elevations. This fog can be maintained in relatively high winds. Usually nocturnal, radiational cooling is also needed to cool the air to its dewpoint. (Great Plains)

B. FRONTAL FOGS

1. <u>Prefrontal (warm front)</u>: rain falling into stable cooler air evaporates and raises the dewpoint of the air to the condensation point. A low-level continental-polar air mass is most favorable for its occurrence as is the presence of a nearby secondary low which produces a weak pressure gradient and light winds. (Mid- and North-Atlantic coast states in Winter; Appalachian valleys.)

2. <u>Post-frontal (cold front)</u>: very little difference from B-1 except that the associated precipitation band is much more restricted in area. The air in both cases must be stable or cumuliform clouds will form. (Midwestern U.S. during polar air mass outbreaks.)

3. <u>Front-passage Fog</u>: a short-lived fog that can form in a variety of ways (mixing of two moist air masses of different temperatures, sudden cooling of air over moist ground, etc.) with the passage of a frontal zone.

C. ICE FOG

Fog caused primarily by man-made sources of moisture (combustion) at temperatures colder than about -30°C. (Polar regions.)

Appendix B - Partial List of Past Warm Fog Modification Efforts and/or Evaluations

General Method	Project Identification or Investigator	Principle Employed
I Evaporation of Fog Droplets	1) Fido	1) Convective Air Heating - Fuel Combustion
	2) J-33 Engines	2) Convective Heating - Fuel Combustion
	3) Telkes	3) Air Heating - Solar Energy
	4) Thermal Res. & Eng. Corp.	4) Heating - Absorption of Radiant Energy by Water Vapor
	5) Houghton - Radford (and various investigators)	5) Air Drying (Chemical Dessication) - $C_a Cl_2$ Droplets
	6) Diesel Power, Inc.	6) Mechanical Mixing with Higher Dry Air
	7) Office of Naval Research	7) Air Heating with Carbon Black
II Growth and Fallout of Fog Droplets	1) Pauthenier	1) Electrical Coalescence of Charged Particles
	2) Cottrell	2) Corona Discharge Charging of Droplets
	3) Plank	3) Cold Water Seeding
	4) Various Investigators	4) Sonic Coagulation of Droplets
III Mechanical Impaction of Fog Droplets	1) Hori	1) Impaction of Droplets on Trees and Foliage
	2) Facy	2) Impaction on Soap Bubbles

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Appendix C - Supercooled Fog Statistics[*]

AVERAGE NUMBER OF HOURS WITH SUPERCOOLED FOG
(based on 3-year period 1957-1960)

Airport	· JAN	FEB	MAR	OCT	NOV	DEC	ANNUAL
Medford, Oregon	56	4	1	3	22	125	213
Spokane, Washington	53	16	0	3	12	76	159
Pendleton, Oregon	29	7	1	0	2	69	107
Boise, Idaho	32	12	0	1	1	26	73
Portland, Oregon	23	5	3	0	12	7	50
Salt Lake City, Utah	19	2	0	0	5	20	46
Seattle, Washington	0	2	0	0	2	9	13
Denver, Colorado	1	4	2	4	1	1	13

NUMBER OF HOURS WITH SUPERCOOLED FOG
MEDFORD

Year	JAN	FEB	MAR	OCT	NOV	DEC	ANNUAL TOTAL
1958	124	3	2	3	19	33	181
1959	29	3	0	2	12	237	293[**]
1960	15	6	0	4	34	105	164
Average	56	4	1	3	22	125	213

3 YEAR DISTRIBUTION BY SPECIFIC TEMPERATURE - MEDFORD

	32°	31°	30°	29°	28°	27°	26°	25°	24°	24°	All
No. of Hours	128	118	120	77	77	54	36	18	1	9	638

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[*]Far West Fog Statistics compiled by United Airlines and supplied through the courtesy of Henry T. Harrison. John F. Kennedy (Idlewild) N. Y. Statistics obtained through the National Weather Records Center.

[**]Including 10 hours in April.

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FOG FREQUENCY - JOHN F. KENNEDY AIRPORT
(Idlewild) New York

MONTH	YEAR	FOG $< 32^{O}F$	FOG $\geq 32^{O}F$	NO FOG $< 32^{O}F$	NO FOG $\geq 32^{O}F$	TOTAL OBS
11	1948		76	1	643	720
	1949		17	66	637	720
	1950		47	6	667	720
	1951		50	77	593	720
	1952		49	15	656	720
	1953		52		668	720
	1954		74		646	720
	1955	1	42	51	626	720
	Total	1	407	216	5136	5760
12	1948	7	56	218	463	744
	1949		112	171	461	744
	1950	1	77	224	442	744
	1951	1	60	231	452	744
	1952	2	53	118	571	744
	1953		58	122	564	744
	1954		48	170	526	744
	1955		32	399	313	744
	Total	11	496	1653	3792	5952
01	1949	4	94	130	516	744
	1950		134	126	484	744
	1951	20	63	223	438	744
	1952		114	226	404	744
	1953	11	135	142	456	744
	1954		40	381	323	744
	1955		12	289	443	744
	1956	4	81	317	342	744
	Total	39	673	1834	3406	5952
02	1949	3	40	143	486	672
	1950	6	67	246	353	672
	1951		70	218	384	672
	1952		24	165	507	696
	1953	2	48	119	503	672
	1954		49	106	517	672
	1955		64	218	390	672
	1956	8	108	166	414	696
	Total	19	470	1381	3554	5424

FOG FREQUENCY - JOHN F. KENNEDY AIRPORT (cont'd)

MONTH	YEAR	FOG <32°F	FOG ≥32°F	NO FOG <32°F	NO FOG ≥32°F	TOTAL OBS
03	1949	4	45	142	553	744
	1950		99	218	427	744
	1951	1	75	54	614	744
	1952		47	76	621	744
	1953		107	100	537	744
	1954		62	96	586	744
	1955	7	110	85	542	744
	1956	7	46	155	536	744
	Total	19	591	926	4416	5952